Figure S1 Hachisako et al.

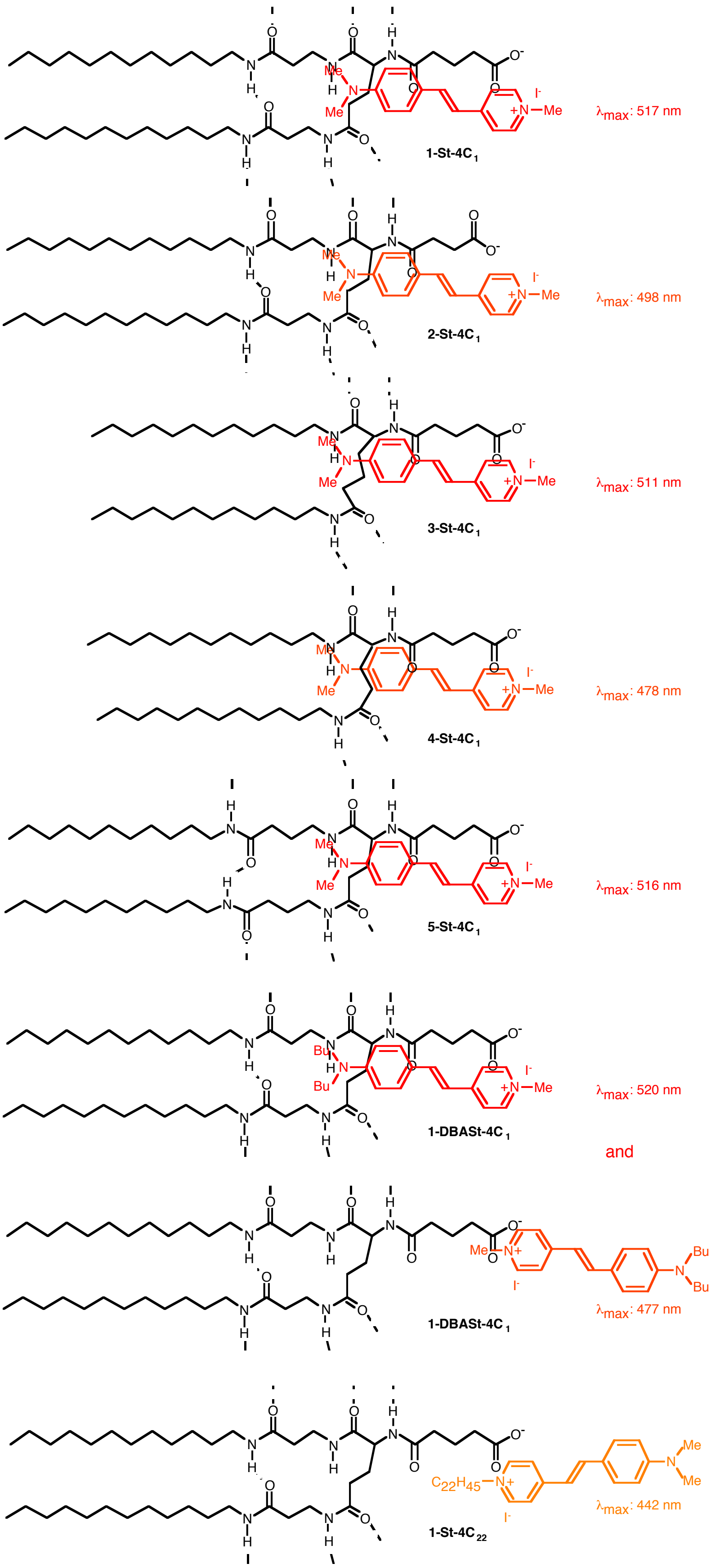

Figure S1. Schematic representation of probable interaction mode between anionic amphiphiles **1**-**5** and representative hemicyanines. It is suggested that β-Ala residues of **1** and GABA residues of **5** are not directly responsible for dye incorporation because electrostatically bound dyes do not reach the β-Ala and GABA moieties of **1** and **5**, respectively, and they rather play crucial role in adjusting packing mode between amphiphiles for formation of specific supramolecular environments (*i.e.*, width and hydrophobicity of cavities) whether they can incorporate hemicyanine dyes specifically or not.

Figure S2 Hachisako et al.

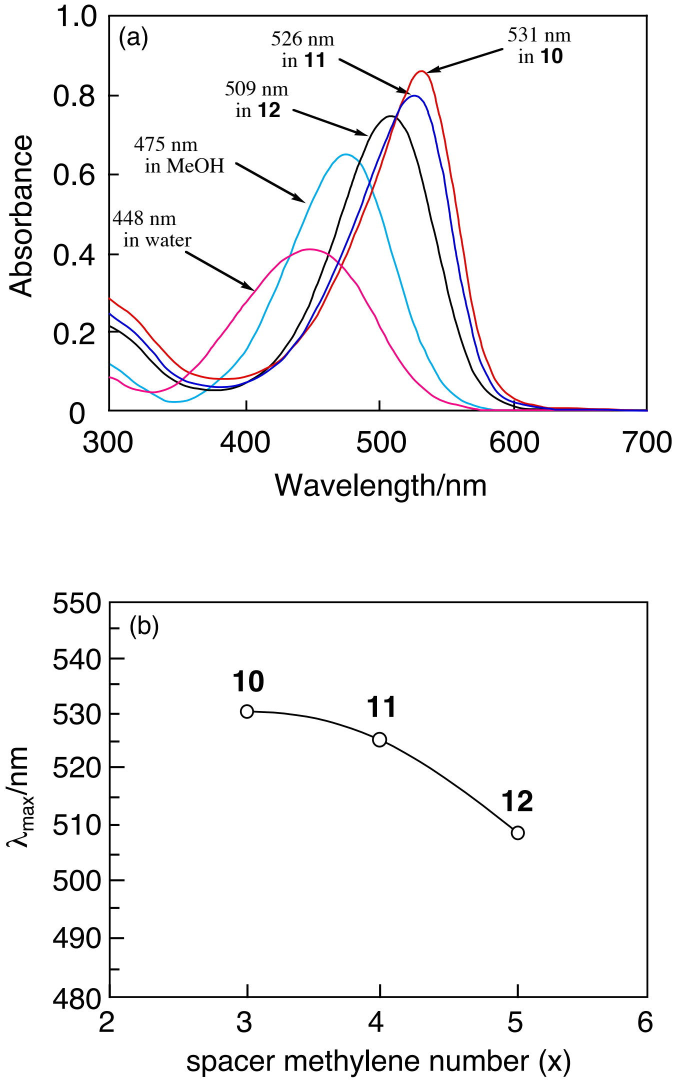

Figure S2. (a) UV-Vis. absorption spectra of aqueous solutions of St-4C_1 in the presence of **10**, **11** and **12**: temperature 20˚C, pH 10, path length: 0.1 cm, [St-4C_1] = 0.15 mM = const., [**10**] = [**11**] = [**12**] = 3.0 mM: (b) Relationships between λ_{max} of St-4C_1 and spacer methylene number of L-Lys-derived amphiphiles **10**, **11** and **12**.

Figure S3 Hachisako et al.

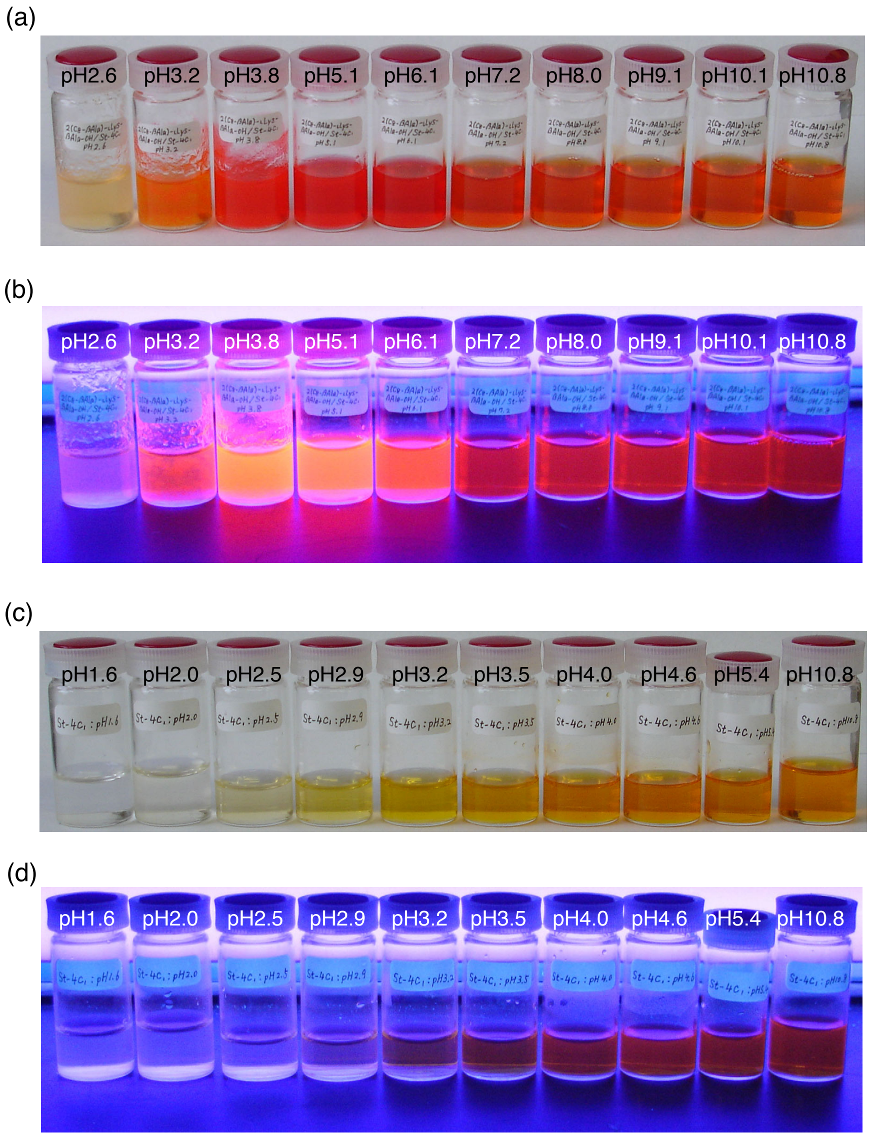

Figure S5. Relationships between pH and the coluor of 0.15 mM of St-4C$_1$ in water in the presence of 3.0 mM of amphiphile **15**; (a) without black light irradiation; (b) under black light irradiation; (c) St-4C$_1$ alone in water without black light irradiation; (d) St-4C$_1$ alone in water under black light irradiation.

Supplementary Material (ESI) for Organic & Biomolecular Chemistry
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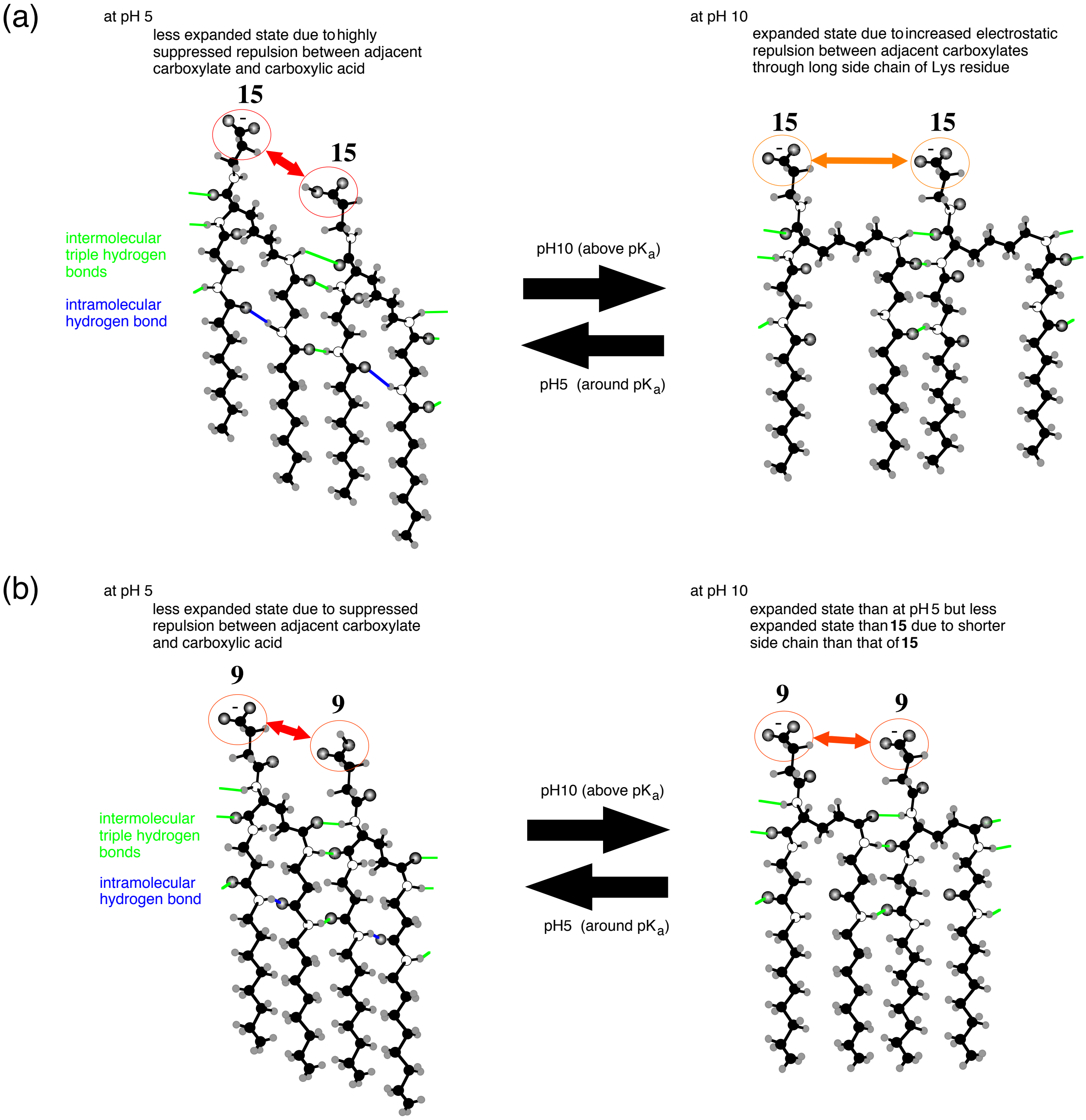

Figure S4. Schematic illustration of pH-dependent expansion and contraction behaviours of L-lysine-derived amphiphile **15** with long side-chain and L-glutamic acid-derived amphiphile **9** with shorter side-chain. Red and orange colours denote the pH-dependent colour change in St-4C_1 incorporated in self-assemblies of **15** and **9**. St-4C_1 electrostatically bound to carboxylate of adjacent amphiphile is omitted for clarity because of [amphiphile]/[St-4C_1] = 20.

Figures S5 and S6 Hachisako et al.

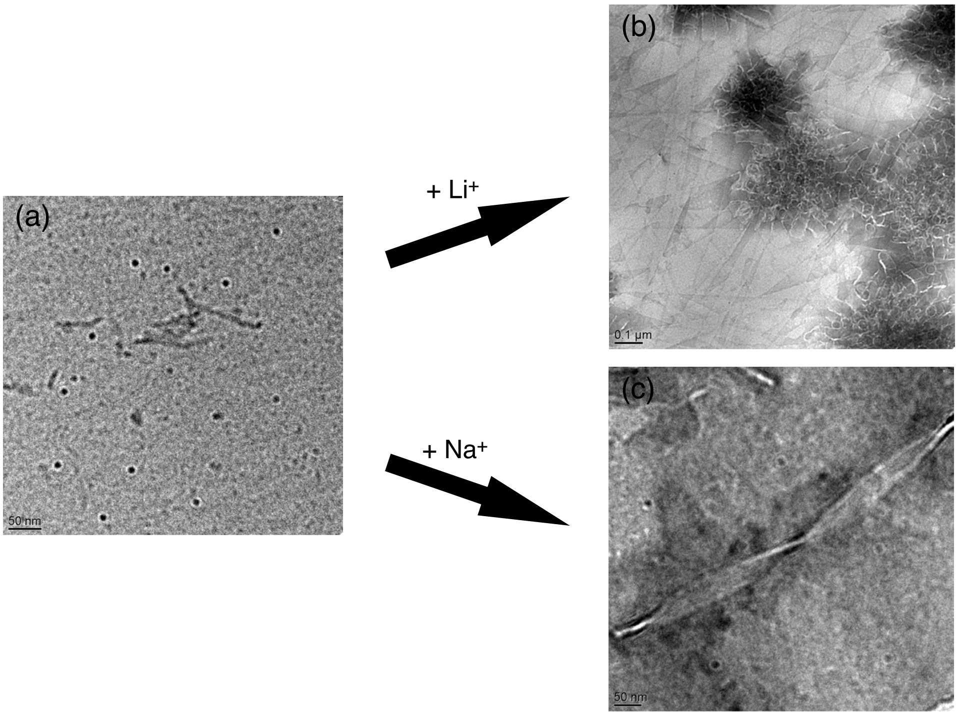

Figure S5. Transmisstion electron micrographs of peptide amphiphile **15** (a) alone in water, and after addition of (b) LiCl and (c) NaCl; pH 10, [**15**] = 3.0 mM, [LiCl] = [NaCl] = 30 mM.

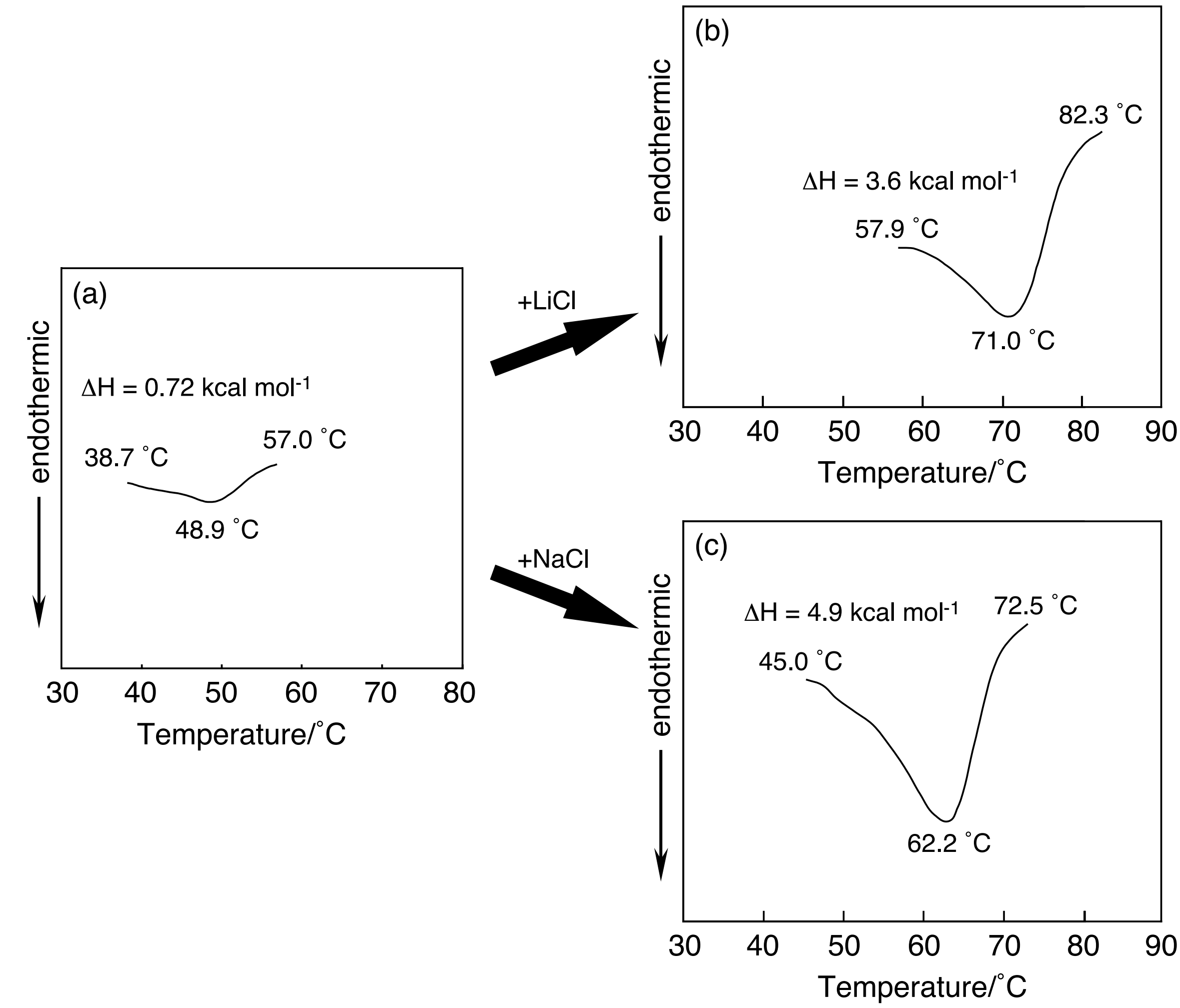

Figure S6. Representive DSC thermograms of peptide amphiphile **15** before (a) and after (b) addition of NaCl; pH 10, [**15**] = 20 mM, [NaCl] = 200 mM, heating rate: 2 ˚C/min.